

09/601356

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TRANSMITTAL LETTER TO THE UNITED STATES

ATTORNEY'S DOCKET NUMBER 48773

DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/EP 99/00578	29 January 1999	11 February 1998

TITLE OF INVENTION: INJECTION STRETCH BLOW MOLDED CONTAINERS MADE FROM OLEFIN POLYMERS

APPLICANT(S) FOR DO/EO/US Bernd Lothar MARCZINKE, Peter John Vaughan JONES, Meinolf KERSTING, David FISCHER, Franz LANGHAUSER, Ulrich MOLL, Wolfgang BIDELE

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. /X/ This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.
 2. / / This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.
 3. /X/ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
 4. /x / A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
 5. /X/ A copy of the International Application as filed (35 U.S.C. 371(c)(2)).
 - a./X/ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b./ / has been transmitted by the International Bureau.
 - c./ / is not required, as the application was filed in the United States Receiving Office (RO/USO).
 6. /X/ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
 7. /X/ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)).
 - a./x/ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b./ / have been transmitted by the International Bureau.
 - c./ / have not been made; however, the time limit for making such amendments has NOT expired.
 - d./ / have not been made and will not be made.
 8. /X/ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
 9. /X/ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
 10. / / A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).
- Items 11. to 16. below concern other document(s) or information included:
11. /X/ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
 12. /X/ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
 13. /X/ A FIRST preliminary amendment.
/ / A SECOND or SUBSEQUENT preliminary amendment.
 14. / / A substitute specification.
 15. / / A change of power of attorney and/or address letter.
 16. /x / Other items or information.
International Search Report
International Preliminary Examination Report

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U.S. Appl. No. (If Known) **09/601356**

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48773

17. /X/ The following fees are submitted	CALCULATIONS	PTO USE ONLY
BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)): Search Report has been prepared by the EPO or JPO.....	\$840.00 840.00	
International preliminary examination fee paid to USPTO (37 CFR 1.482).....	\$750.00	
No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)).....	\$700.00	
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO	\$ 970.00	
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied pro -visions of PCT Article 33(2)-(4).....	\$96.00	

ENTER APPROPRIATE BASIC FEE AMOUNT = \$ 840.00

Surcharge of \$130.00 for furnishing the oath or declaration later than / / 20 / 30 months from the earliest claimed priority date (37 CFR 1.492(e)).

<u>Claims</u>	<u>Number Filed</u>	<u>Number Extra</u>	<u>Rate</u>
Total Claims	6 -20		X\$18.
Indep.Claims	1 -3		X\$78.
Multiple dependent claim(s)(if applicable)			+270.
TOTAL OF ABOVE CALCULATION			= 840.00

Reduction of 1/2 for filing by small entity, if applicable.
Verified Small Entity statement must also be filed
(Note 37 CFR 1.9, 1.27, 1.28).

SUBTOTAL	=	840.00
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Processing fee of \$130. for furnishing the English translation later than / /20 / /30 months from the earliest claimed priority date (37 CFR 1.492(f)). +

TOTAL NATIONAL FEE	=	840.00
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Fee for recording the enclosed assignment (37 CFR 1.21(h)).
The assignment must be accompanied by an appropriate cover
sheet (37 CFR 3.28, 3.31) \$40.00 per property =

TOTAL FEES ENCLOSED	=	\$	840.00 880.00
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Amount to be

refunded: \$

Charged	\$
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a./X/ A check in the amount of \$ 880. ~~240.~~ to cover the above fees is enclosed.

b./ / Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c./X/ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 11-0345. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:
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Herbert B. Keil
NAME
18,967
Registration No.

SIGNATURE

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Parameter	Value	Unit
Initial concentration	1.0	g/L
Initial pH	7.0	
Temperature	25	°C
Time	0, 1, 2, 4, 8, 16, 32, 64, 128, 256, 512, 1024, 2048, 4096, 8192, 16384, 32768, 65536, 131072, 262144, 524288, 1048576, 2097152, 4194304, 8388608, 16777216, 33554432, 67108864, 134217728, 268435456, 536870912, 1073741824, 2147483648, 4294967296, 8589934592, 17179869184, 34359738368, 68719476736, 137438953472, 274877906944, 549755813888, 1099511627776, 2199023255552, 4398046511104, 8796093022208, 17592186044416, 35184372088832, 70368744177664, 140737488355328, 281474976710656, 562949953421312, 1125899906842624, 2251799813685248, 4503599627370496, 9007199254740992, 18014398509481984, 36028797018963968, 72057594037927936, 144115188075855872, 288230376151711744, 576460752303423488, 1152921504606846976, 2305843009213693952, 4611686018427387904, 9223372036854775808, 18446744073709551616, 36893488147419103232, 73786976294838206464, 147573952589676412928, 295147905179352825856, 590295810358705651712, 1180591620717411303424, 2361183241434822606848, 4722366482869645213696, 9444732965739290427392, 18889465931478580854784, 37778931862957161709568, 75557863725914323419136, 151115727451828646838272, 302231454903657293676544, 604462909807314587353088, 1208925819614629174706176, 2417851639229258349412352, 4835703278458516698824704, 9671406556917033397649408, 19342813113834066795298816, 38685626227668133590597632, 77371252455336267181195264, 154742504910672534362390528, 309485009821345068724781056, 618970019642690137449562112, 1237940039285380274899124224, 2475880078570760549798248448, 4951760157141521099596496896, 9903520314283042199192993792, 19807040628566084398385987584, 39614081257132168796771975168, 79228162514264337593543950336, 158456325028528675187087900672, 316912650057057350374175801344, 633825300114114700748351602688, 1267650600228229401496703205376, 2535301200456458802993406410752, 5070602400912917605986812821504, 10141204801825835211973625643008, 20282409603651670423947251286016, 40564819207303340847894502572032, 81129638414606681695789005144064, 162259276829213363391578010288128, 324518553658426726783156020576256, 649037107316853453566312041152512, 1298074214633706907132624082305024, 2596148429267413814265248164610048, 5192296858534827628530496329220096, 10384593717069655257060992658440192, 20769187434139310514121985316880384, 41538374868278621028243970633760768, 83076749736557242056487941267521536, 166153499473114484112975882535043072, 332306998946228968225951765070086144, 664613997892457936451903530140172288, 1329227995784915872903807060280344576, 2658455991569831745807614120560689152, 5316911983139663491615228241121378304, 10633823966279326983230456482242756608, 21267647932558653966460912964485513216, 42535295865117307932921825928971026432, 85070591730234615865843651857942052864, 170141183460469231731687303715884105728, 340282366920938463463374607431768211456, 680564733841876926926749214863536422912, 1361129467683753853853498429727072845824, 2722258935367507707706996859454145691648, 5444517870735015415413993718908291383296, 10889035741470030830827987437816582766592, 21778071482940061661655974875633165533184, 43556142965880123323311949751266331066368, 87112285931760246646623899502532662132736, 174224571863520493293247799005065324265472, 348449143727040986586495598010130648530944, 696898287454081973172991196020261297061888, 1393796574908163946345982392040522594123776, 2787593149816327892691964784081045188247552, 5575186299632655785383929568162090376495104, 11150372599265311570767859136324180752990208, 22300745198530623141535718272648361505980416, 44601490397061246283071436545296723011960832, 89202980794122492566142873090593446023921664, 178405961588244985132285746181186892047843328, 356811923176489970264571492362373784095686656, 713623846352979940529142984724747568191373312, 1427247692705959881058285969449495136382746624, 2854495385411919762116571938898990272765493248, 5708990770823839524233143877797980545530986496, 11417981541647679048466287755595961091061972992, 2283596308329	

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Injection stretch blow molded containers made from olefin polymers

- 5 The present invention relates to injection stretch blow molded containers made from olefin polymers, comprising homopolymers of propylene or copolymers of propylene with other C₂-C₁₀-alk-1-enes, which are obtainable by polymerizing the corresponding monomers with metallocene catalysts. The invention also relates to
- 10 processes for producing injection stretch blow molded containers from olefin polymers and to the use of olefin polymers for producing injection stretch blow molded containers.

- Due to their excellent resistance to heat and chemicals,
- 15 exceptionally good stiffness and toughness, and also their cost-effectiveness, containers and bottles made from olefin polymers are widely used, for example as packaging for food and drink, household items or cosmetics, and in the medical sector. Injection stretch blow molding (ISBM) has been developed in order
- 20 to obtain containers of adequate transparency.

- Injection stretch blow molding has been known for quite some time. US-A 4 357 288 teaches a process in which initially a parison is injection molded from a crystalline polypropylene at a
- 25 temperature which is only slightly higher than the lowest temperature at which a clear melt is obtained, and the parison is then cooled until it hardens. The parison is then heated again to a temperature just below the amorphous flow temperature and stretch blow molded.

- 30 EP-A 151 741 describes a process which includes three steps for producing containers from propylene polymers, in which initially a parison is injection molded. The significant step is then that this is preblown in a specific temperature range; only after this
- 35 has been done is it stretch blow molded. Suitable polymers are propylene copolymers with a comonomer content of from 1 to 6% by weight and a melt flow rate of from 4 to 50 g/10 min. This text also teaches that amounts of up to 1% by weight of nucleating agents, such as dibenzylidenesorbitol or a derivative thereof,
- 40 may be added to the polymers to improve transparency.

- EP-A 309 138 relates to a process for producing containers from propylene copolymers which uses propylene-ethylene copolymers with an ethylene content of from 0.5 to 8% by weight and having a
- 45 melt flow rate of greater than 50 g/min. The preblowing step is not essential as long as the parison produced by injection molding in the first step is in a partially molten state when it

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- H. Heyn, in Kunststoffe 84, 1994, pages 1415 - 1418 describes a two-step injection stretch blow molding process in which polypropylene parisons which have been separately injection molded are processed after they have been heated once again, to give hollow articles.

- It is an object of the present invention to set aside the disadvantages described and in particular to provide containers which have excellent transparency combined with good mechanical properties and small contents of soluble fractions, and which can be produced by a process which has a wide temperature window and permits short cycle times.

- 45 We have found that this object is achieved by means of injection stretch blow molded containers made from olefin polymers, comprising homopolymers of propylene or copolymers of propylene

with other C₂-C₁₀-alk-1-enes, which can be obtained by polymerizing the corresponding monomers with metallocene catalysts, processes for producing injection stretch blow molded containers from olefin polymers and the use of olefin polymers for producing
5 injection stretch blow molded containers.

For the purposes of the invention, olefin polymers are homo- or copolymers of linear or branched α olefins, in particular of C₂-C₁₀-alk-1-enes, such as ethylene, propylene, 1-butene,
10 1-pentene, 1-hexene, 1-heptene, 1-octene, 1-nonene, 1-decene or 4-methyl-1-pentene, or mixtures of these α -olefins. Preference is given to homo- or copolymers of ethylene or of propylene, where the proportion of ethylene or of propylene in the copolymers is at least 50 mol%. Among the copolymers of ethylene, preference is
15 given to those which comprise, as other monomers, propylene, 1-butene, 1-hexene or 1-octene, or mixtures of these. The copolymers of propylene are in particular those which comprise, as other monomers, ethylene or 1-butene, or mixtures of these. The propylene copolymers may comprise the other α olefins
20 incorporated randomly. They may also take the form of block copolymers or impact copolymers. Preferred polymers of propylene are propylene homopolymers and random propylene copolymers. Olefin polymers of this type are known per se or may be prepared by known methods by polymerizing with conventional Ziegler-Natta
25 catalysts or with metallocene catalysts.

The olefin polymers of which the novel injection stretch blow molded containers are composed comprise propylene polymers, which are obtainable by polymerizing the corresponding monomers with
30 metallocene catalysts. The proportion of the propylene polymers which are obtainable with metallocene catalysts is generally at least 40% by weight, preferably at least 70% by weight and in particular at least 90% by weight, based on the olefin polymers. In a very particularly preferred embodiment, the olefin polymers
35 are obtained by homopolymerization of propylene or copolymerization of propylene with other C₂-C₁₀-alk-1-enes with metallocene catalysts.

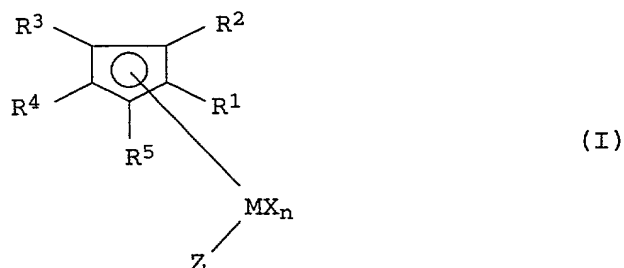
The propylene polymers which are obtainable with metallocene
40 catalysts are propylene homopolymers or copolymers made from propylene and another C₂-C₁₀-alk-1-ene or more than one other C₂-C₁₀-alk-1-ene, preferably selected from the class consisting of ethylene, 1-butene, 1-pentene, 1-hexene, 1-heptene, 1-octene, 1-nonene, 1-decene and 4-methyl-1-pentene, or are mixtures of
45 these polymers, in which case the mixing ratios are not critical. The copolymers of propylene used are generally random copolymers.

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For the purposes of the invention, metallocene catalysts are substances which are generally obtained by combining a transition metal compound or more than one transition metal compound, preferably of titanium, of zirconium or of hafnium, and comprising at least one ligand which, in the broadest sense, is a derivative of the cyclopentadienyl ligand, with an activator, also called cocatalyst or metallocenium-ion-forming compound, and generally have activity for polymerizing the monomers described. Examples of such catalysts are described in EP-A 545 303, EP-A 576 970 and EP-A 582 194. Metallocene catalysts which are suitable according to the invention generally comprise, as active constituents,

A) one or more metallocene complexes of the general formula (I)



where:

25 M is titanium, zirconium, hafnium, vanadium, niobium or tantalum, or also elements of the 3rd subgroup of the Periodic Table or of the lanthanoides,

30 X is fluorine, chlorine, bromine, iodine, hydrogen, C₁-C₁₀-alkyl, C₆-C₁₅-aryl, alkylaryl having from 1 to 10 carbon atoms in the alkyl radical and from 6 to 20 carbon atoms in the aryl radical, -OR⁶ or -NR⁶R⁷,

35 n is 1, 2 or 3, where n is the valence of M minus the number 2,

where

40 R⁶ and R⁷ are C₁-C₁₀-alkyl, C₆-C₁₅-aryl, alkylaryl, arylalkyl, fluoroalkyl or fluoroaryl having in each case from 1 to 10 carbon atoms in the alkyl radical and from 6 to 20 carbon atoms in the aryl radical, and

45 the radicals X are identical or different,

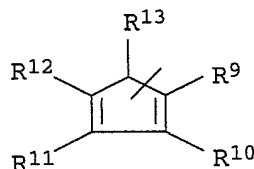
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R^1 to R^5 are hydrogen, C_1 - C_{10} -alkyl, 5- to 7-membered cycloalkyl, which in turn may be C_1 - C_{10} -alkyl-substituted, C_6 - C_{15} -aryl or arylalkyl, where two adjacent radicals together may also be saturated or unsaturated cyclic groups having from 4 to 15 carbon atoms, or $Si(R^8)_3$, where

R^8 may be C_1 - C_{10} -alkyl, C_3 - C_{10} -cycloalkyl or C_6 - C_{15} -aryl, and

10

Z is X or



15

where

R^9 to R^{13} are hydrogen, C_1 - C_{10} -alkyl, 5- to 7-membered cycloalkyl, which in turn may be C_1 - C_{10} -alkyl-substituted, C_6 - C_{15} -aryl or arylalkyl, where two adjacent radicals together may also be saturated or unsaturated cyclic groups having from 4 to 15 carbon atoms, or $Si(R^{14})_3$, where

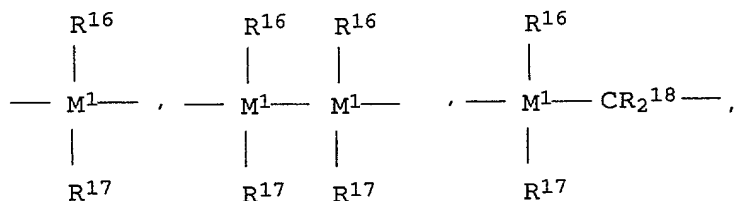
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R^{14} is C_1 - C_{10} -alkyl, C_3 - C_{10} -cycloalkyl or C_6 - C_{15} -aryl,

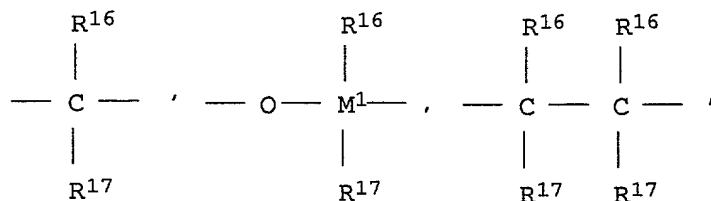
or where R^4 and Z together form a grouping $-R^{15}-A-$, where

30

R^{15} is



35



40

$= BR^{16}$, $= AlR^{16}$, $-Ge-$, $-Sn-$, $-O-$, $-S-$, $= SO$, $= SO_2$,
 $= NR^{16}$, $= CO$, $= PR^{16}$ or $= P(O)R^{16}$,

45

where

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each of R^{16} , R^{17} and R^{18} is identical or different and is hydrogen, halogen, C_1 - C_{10} -alkyl, C_1 - C_{10} -fluoro-alkyl, C_6 - C_{10} -fluoroaryl, C_6 - C_{10} -aryl, C_1 - C_{10} -alkoxy, C_2 - C_{10} -alkenyl, C_7 - C_{40} -arylalkyl, C_8 - C_{40} -arylalkenyl or C_7 - C_{40} -alkylaryl, or where two adjacent radicals, in each case with the atoms linking them, form a saturated or unsaturated ring having from 4 to 15 carbon atoms, and

10 M^1 is silicon, germanium or tin,

A is $—O—$, $—S—$, >NR^{19} or >PR^{19} ,

where

15

R^{19} is C_1 - C_{10} -alkyl, C_6 - C_{15} -aryl, C_3 - C_{10} -cycloalkyl, C_7 - C_{18} -alkylaryl or $\text{Si}(R^{20})_3$,

20

R^{20} is hydrogen, C_1 - C_{10} -alkyl, C_6 - C_{15} -aryl, which may in turn be substituted with C_1 - C_4 -alkyl groups, or is C_3 - C_{10} -cycloalkyl,

or where R^4 and R^{12} together form a grouping $-R^{15}-$,

25

and

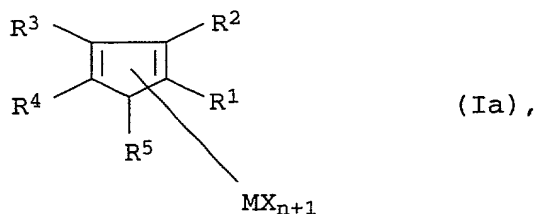
B) one or more metallocenium-ion-forming compounds.

30

The radicals X in the general formula (I) are preferably identical.

Among the metallocene complexes of the general formula (I), preference is given to

35



40

45

5



M is titanium, zirconium or hafnium,
40 X is chlorine, C₁-C₄-alkyl or phenyl,
n is the number 2 and

45

8

Among the compounds of the formula (Ib), preference is given to those in which

- M is titanium, zirconium or hafnium,
5 X is chlorine, C₁-alkyl or phenyl,
n is the number 2,
10 R¹ to R⁵ are hydrogen, C₁-C₄-alkyl or Si(R⁸)₃ and
R⁹ to R¹³ are hydrogen, C₁-C₄-alkyl or Si(R¹⁴)₃.

Particularly suitable compounds of the formula (Ib) are those in
15 which the cyclopentadienyl radicals are identical.

- Examples of particularly suitable compounds are:
bis(cyclopentadienyl)zirconium dichloride,
bis(pentamethylcyclopentadienyl)zirconium dichloride,
20 bis(methylcyclopentadienyl)zirconium dichloride,
bis(ethylcyclopentadienyl)zirconium dichloride,
bis(n-butylcyclopentadienyl)zirconium dichloride and
bis(trimethylsilylcyclopentadienyl)zirconium dichloride,
and also the corresponding dimethylzirconium compounds.
25 Particularly suitable compounds of the formula (Ic) are those in
which

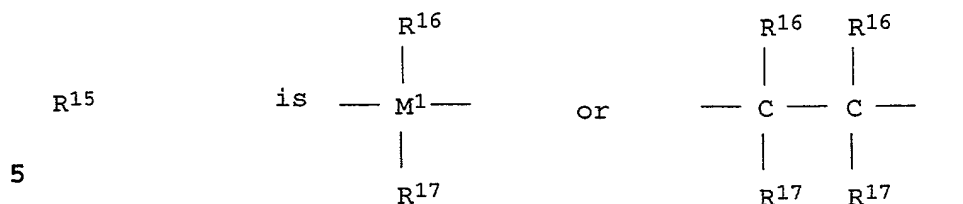
- R¹ and R⁹ are identical and are hydrogen or C₁-C₁₀-alkyl,
30 R⁵ and R¹³ are identical and are hydrogen, methyl, ethyl,
isopropyl or tert-butyl,
R³ and R¹¹ are C₁-C₄-alkyl and
35 R² and R¹⁰ are hydrogen

or

- two adjacent radicals R² and R³, or also R¹⁰ and R¹¹, together are
40 a saturated or unsaturated cyclic group having from 4 to 12
carbon atoms,

45

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M is titanium, zirconium or hafnium, and

10 X is chlorine, C₁-C₄-alkyl or phenyl.

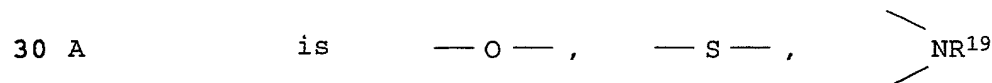
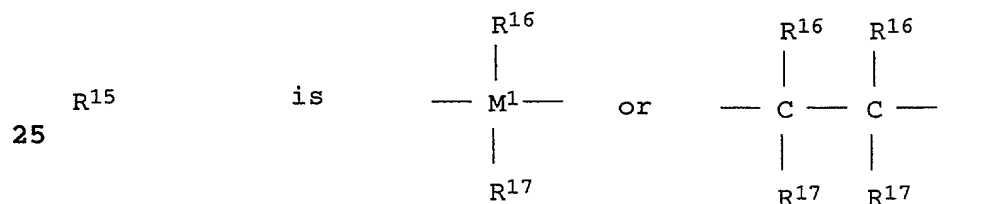
- Examples of particularly suitable complex compounds are
 dimethylsilanediylbis(cyclopentadienyl)zirconium dichloride,
 dimethylsilanediylbis(indenyl)zirconium dichloride,
 15 dimethylsilanediylbis(tetrahydroindenyl)zirconium dichloride,
 ethylenebis(cyclopentadienyl)zirconium dichloride,
 ethylenebis(indenyl)zirconium dichloride,
 ethylenebis(tetrahydroindenyl)zirconium dichloride,
 tetramethylethylene-9-fluorenylcyclopentadienylzirconium
 20 dichloride,
 dimethylsilanediylbis(3-tert-butyl-5-methylcyclopentadienyl)-
 zirconium dichloride,
 dimethylsilanediylbis(3-tert-butyl-5-ethylcyclopentadienyl)-
 zirconium dichloride,
 25 dimethylsilanediylbis(2-methylindenyl)zirconium dichloride,
 dimethylsilanediylbis(2-isopropylindenyl)zirconium dichloride,
 dimethylsilanediylbis(2-tert-butylindenyl)zirconium dichloride,
 diethylsilanediylbis(2-methylindenyl)zirconium dibromide,
 dimethylsilanediylbis(3-methyl-5-methylcyclopentadienyl)-
 30 zirconium dichloride,
 dimethylsilanediylbis(3-ethyl-5-isopropylcyclopentadienyl)zir-
 conium dichloride,
 dimethylsilanediylbis(2-ethylindenyl)zirconium dichloride,
 dimethylsilanediylbis[3,3'-(2-methylbenzindenyl)]zirconium
 35 dichloride,
 dimethylsilanediylbis[3,3'-(2-ethylbenzindenyl)]zirconium
 dichloride,
 methylphenylsilanediylbis[3,3'-(2-ethylbenzindenyl)]zirconium
 dichloride,
 40 methylphenylsilanediylbis[3,3'-(2-methylbenzindenyl)]zirconium
 dichloride,
 diphenylsilanediylbis[3,3'-(2-methylbenzindenyl)]zirconium
 dichloride,
 diphenylsilanediylbis[3,3'-(2-ethylbenzindenyl)]zirconium
 45 dichloride, and

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5 dimethylsilanediylbis(2-methyl-4-phenylindenyl)zirconium
dichloride,
dimethylsilanediylbis(2-methyl-4-naphthylindenyl)zirconium
dichloride,
dimethylsilanediylbis(2-methyl-4-isopropylindenyl)zirconium
10 dichloride, and
dimethylsilanediylbis(2-methyl-4,6-diisopropylindenyl)zirconium
dichloride, and also the corresponding dimethylzirconium
compounds.

M is titanium or zirconium,

20 X is chlorine, C₁-C₄-alkyl or phenyl.



and

R¹ to R³ and R⁵ are hydrogen, C₁-C₁₀-alkyl, C₃-C₁₀-cycloalkyl,
35 C₆-C₁₅-aryl or Si(R⁸)₃, or where two adjacent radicals
are a cyclic group having from 4 to 12 carbon atoms.

Complex compounds of this type may be synthesized by methods known per se. Preference is given to reacting the corresponding substituted cyclic hydrocarbon anions with halides of titanium, zirconium, hafnium, vanadium, niobium or tantalum.

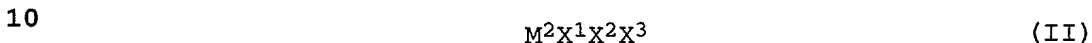
Examples of corresponding preparation processes are described, inter alia, in the Journal of Organometallic Chemistry, 369 45 (1989), 359-370.

11

It is also possible to use mixtures of different metallocene complexes.

Examples of suitable metallocenium-ion-forming compounds B) are
 5 strong neutral Lewis acids, ionic compounds with Lewis-acid cations and ionic compounds with Brönsted acids as cations.

Preferred strong, neutral Lewis acids are compounds of the general formula (II)



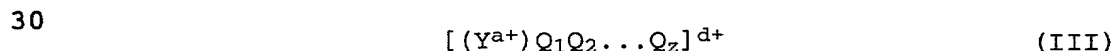
where

15 M^2 is an element of the 3rd main group of the Periodic Table, in particular B, Al or Ga, preferably B, and

20 X^1 , X^2 and X^3 are hydrogen, C_1 - C_{10} -alkyl, C_6 - C_{15} -aryl, alkylaryl, arylalkyl, haloalkyl or haloaryl with in each case from 1 to 10 carbon atoms in the alkyl radical and from 6 to 20 carbon atoms in the aryl radical or fluorine, chlorine, bromine or iodine, in particular haloaryl, preferably pentafluorophenyl.

25 Particular preference is given to compounds of the general formula (II), in which X^1 , X^2 and X^3 are identical, preferably tris(pentafluorophenyl)borane.

Suitable ionic compounds with Lewis-acid cations are compounds of the general formula (III)



where

35 Y is an element of the 1st to 6th main group or of the 1st to 8th subgroup of the Periodic Table,

40 Q_1 to Q_z are radicals with a single negative charge, such as C_1 - C_{28} -alkyl, C_6 - C_{15} -aryl, alkylaryl, arylalkyl, haloalkyl, haloaryl with in each case from 6 to 20 carbon atoms in the aryl radical and from 1 to 28 carbon atoms in the alkyl radical, C_3 - C_{10} -cycloalkyl, which may be unsubstituted or substituted with C_1 - C_{10} -alkyl groups, or are halogen, C_1 - C_{28} -alkoxy, C_6 - C_{15} -aryloxy, silyl or mercaptyl,

45 a is an integer from 1 to 6,

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z is an integer from 0 to 5, and

d is the difference a-z, and is greater than or equal to 1.

5

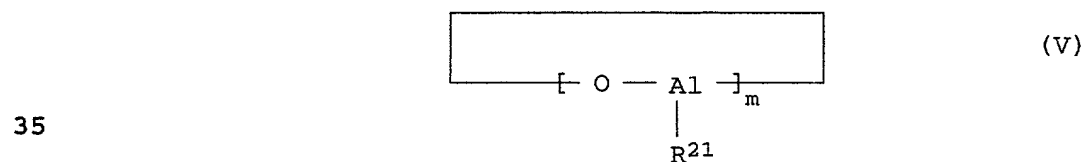
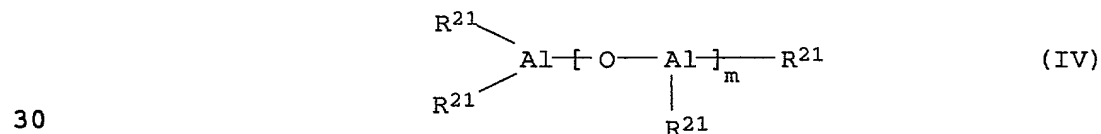
Particularly suitable cations are carbonium cations, oxonium cations and sulfonium cations, and also cationic transition metal complexes. The triphenylmethyl cation, the silver cation and the 1,1'-dimethylferrocenyl cation should be mentioned in particular.

10 They preferably have non-coordinating counterions, in particular boron compounds, as also mentioned in WO 91/09882, preferably tetrakis(pentafluorophenyl) borate.

Ionic compounds with Brönsted acids as cations and preferably
15 likewise non-coordinating counterions are mentioned in WO 91/09882, and a preferred cation is N,N-dimethylanilinium.

The amount of strong, neutral Lewis acids, of ionic compounds with Lewis-acid cations or of ionic compounds with Brönsted acids
20 as cations is preferably from 0.1 to 10 equivalents, based on the metallocene complex A).

Particularly suitable metallocenium-ion-forming compounds B) are open-chain or cyclic aluminoxane compounds of the general
25 formulae (IV) or (V)



where R^{21} is C_1 - C_4 -alkyl, preferably methyl or ethyl, and m is
40 an integer from 5 to 30, preferably from 10 to 25.

These oligomeric aluminoxane compounds are generally prepared by reacting a solution of trialkylaluminum with water, as described, inter alia, in EP-A 284 708 and US-A 4 794 096.

45 The oligomeric aluminoxane compounds thus obtained are generally mixtures of different linear and/or cyclic long-chain molecules, and m is therefore to be regarded as an average value. The

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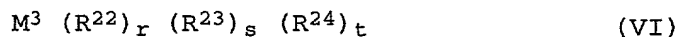
aluminoxane compounds may also be present in a mixture with other metal alkyl compounds, preferably with alkylaluminum compounds.

It has proven advantageous to use amounts of the metallocene complexes A) and of the oligomeric aluminoxane compounds of the general formulae (IV) or (V) which give an atomic ratio of aluminum from the oligomeric aluminoxane compounds to transition metal from the metallocene complexes in the range from 10:1 to 10⁶:1, in particular from 10:1 to 10⁴:1.

It is moreover possible for the component B) used to be aryloxyaluminoxanes, as described in US-A 5 391 793, aminoaluminoxanes, as described in US-A 5 371 260, aminoaluminoxane hydrochlorides, as described in EP-A 633 264, siloxyaluminoxanes, as described in EP-A 621 279, or mixtures of these, instead of the aluminoxane compounds of the general formulae (IV) or (V).

The metallocene complexes A), and also the metallocenium-ion-forming compounds B), are preferably used in solution, particular preference being given to aromatic hydrocarbons having from 6 to 20 carbon atoms, in particular xylenes and toluene.

Suitable metallocene catalysts may also comprise, as further component C), a metallic compound of the general formula (VI)



where

- M^3 is an alkali metal, an alkaline-earth metal or a metal of the 3rd main group of the Periodic Table i.e. boron, aluminum, gallium, indium or thallium,
- R^{22} is hydrogen, C₁-C₁₀-alkyl, C₆-C₁₅-aryl, alkylaryl or arylalkyl having in each case from 1 to 10 carbon atoms in the alkyl radical and from 6 to 20 carbon atoms in the aryl radical,
- R^{23} and R^{24} are hydrogen, halogen, C₁-C₁₀-alkyl, C₆-C₁₅-aryl, alkylaryl, arylalkyl or alkoxy having in each case from 1 to 10 carbon atoms in the alkyl radical and from 6 to 20 carbon atoms in the aryl radical,
- r is an integer from 1 to 3,

5

10

R²³ and R²⁴ are C₁-C₁₀-alkyl.

The metallocene complexes A) may also be used on a support material.

The support materials used are preferably finely divided supports which generally have a particle diameter in the range from 1 to 300 μm , in particular from 20 to 90 μm . Examples of suitable support materials are inorganic oxides of silicon, of aluminum, of titanium, or of one of the metals of the 1st or 2nd main group of the Periodic Table, among which, besides alumina and magnesium oxide and phyllosilicates, preference is in particular given to silica gel.

45 Examples of other supports are finely divided polyolefins, for
example finely divided polypropylene.

The propylene polymers obtainable with metallocene catalysts may be prepared, either batchwise or preferably continuously, in the reactors which are usual for polymerizing olefins. Examples of suitable reactors are continuously-operated stirred tank

5 reactors, and it is also possible, if desired, to use a series of more than one stirred tank reactor connected in series. The polymerization reactions may be carried out in the gas phase, in suspension, in liquid monomers, in supercritical monomers, or in inert solvents.

10

The polymerization conditions are not critical per se. Pressures of from 1 to 3500 bar, preferably from 2 to 100 bar have proven suitable, as have temperatures of from 0 to 400°C, preferably from 20 to 250°C and in particular from 50 to 100°C.

15

The homopolymerizations of propylene or the copolymerizations of propylene with C₂-C₁₀-alk-1-enes are preferably carried out in the gas phase, for example in fluidized-bed reactors or in agitated powder bed reactors. Polymerization conditions well suited for

20 this are polymerization pressures in the range from 10 to 40 bar and polymerization temperatures in the range from 50 to 100°C. The polymerization may also, of course, take place in a series of more than one, preferably two, reactors connected to one another in series.

25

The average molar mass of the polymers may be controlled using the methods usual in polymerization technology, for example by introducing molar mass regulators, such as hydrogen, which gives a reduction in the molar mass of the polymer, or by varying the

30 polymerization temperature. High polymerization temperatures likewise usually give reduced molar masses.

The homopolymers of propylene or the copolymers of propylene with C₂-C₁₀-alk-1-enes, obtainable with metallocene catalysts,

35 generally have a melt flow rate (MFR), measured at 230°C under a weight of 2.16 kg in accordance with ISO 1133, in the range from 0.1 to 1000 g/10 min, preferably from 0.5 to 200 g/10 min, in particular from 1 to 100 g/10 min, and very particularly preferably from 2 to 50 g/10 min. Melt flow rates of this type

40 usually correspond to average molar masses (weight average M_w) of the propylene polymers, measured by the gel permeation chromatography (GPC) method at 140°C in 1,2,4-trichlorobenzene against a polypropylene standard, in the range from 50,000 to 1,000,000 g/mol.

45

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The molar mass distribution M_w/M_n of the homopolymers of propylene or of the copolymers of propylene with C_2-C_{10} -alk-1-enes, obtainable with metallocene catalysts, determined using GPC as for the average molar mass M_w , is generally in the range from 1.2 to 6.0, preferably from 1.5 to 3.0.

Both the molar mass M_w and the molar mass distribution M_w/M_n , and also in particular the MFR, may also be adjusted by the method of peroxidically initiated degradation of a starting polymer, advantageously in an extruder. This method is known to the person skilled in the art.

The propylene homopolymers obtainable with metallocene catalysts generally have a melting point, determined by the differential scanning calorimetry (DSC) method (heating rate $20^\circ\text{C}/\text{min}$), in the range from 80 to 165°C , preferably from 100 to 165°C and in particular from 120 to 165°C .

The copolymers of propylene with other C_2-C_{10} -alk-1-enes generally have a melting point determined by DSC (heating rate $20^\circ\text{C}/\text{min}$) in the range from 60 to 160°C , preferably from 100 to 160°C and in particular from 120 to 160°C .

The proportion of chemically bonded comonomer in the copolymers of propylene with other C_2-C_{10} -alk-1-enes, measured by ^{13}C NMR spectroscopy, for example, is generally in the range from 0.001 to 35 mol%, preferably from 0.01 to 15 mol%, based on the copolymer.

Propylene homopolymers or copolymers of propylene with other C_2-C_{10} -alk-1-enes with very good suitability according to the invention are obtained if the corresponding monomers are polymerized in the presence of a metallocene catalyst which comprises dimethylsilanediylbis[3,3'-(2-methylbenzindenyl)]-zirconium dichloride as metallocene complex (I).

The propylene homopolymers or propylene copolymers obtained in this way then generally have the polymer properties already mentioned above, such as MFR, M_w , M_w/M_n , melting point, pentad content and comonomer content.

Preferred copolymers of propylene with other C_2-C_{10} -alk-1-enes obtainable with the dimethylsilanediylbis[3,3'-(2-methylbenzindenyl)]zirconium dichloride catalyst are random copolymers of propylene and ethylene with, based on the polymer, from 0.1 to 10 mol%, preferably from 0.2 to 7 mol%, of units derived from ethylene, or are random copolymers of propylene and 1-butene

with, based on the polymer, from 0.5 to 20 mol%, preferably from 1 to 15 mol%, of units derived from 1-butene, or are random terpolymers of propylene, ethylene and 1-butene with, based on the terpolymer, from 0.1 to 10 mol%, preferably from 0.2 to 7 5 mol%, of units derived from ethylene and, based on the terpolymer, from 0.5 to 20 mol%, preferably from 1 to 15 mol%, of units derived from 1-butene.

The olefin polymers suitable for producing the novel injection 10 stretch blow molded containers may comprise additives such as stabilizers, lubricants, mold-release agents, fillers, antistats, plasticizers, dyes, pigments, flame retardants or nucleating agents, which are added in the usual amounts before use. These are generally incorporated into the polymer during pelletizing of 15 the polymerization product, which is produced in the form of a powder.

Additives of this type are generally commercially available and are described, for example, in Gächter/Müller, *Plastics Additives* 20 *Handbook*, 4th Edition, Hansa Publishers, Munich, 1993.

Usual stabilizers are antioxidants, such as sterically hindered phenols, process stabilizers, such as phosphites or phosphonites, neutralizing agents, such as calcium stearate or zinc stearate or 25 dihydrotalcite, sterically hindered amines, or else UV stabilizers. The polymers according to the invention generally comprise one or more of the stabilizers in amounts of up to 2% by weight.

Examples of suitable lubricants and mold-release agents are fatty 30 acids, calcium salts of the fatty acids, zinc salts of the fatty acids, fatty amides and low-molecular-weight polyolefin waxes, which are usually used in concentrations of up to 2% by weight.

Examples of possible fillers which may be used for the random 35 propylene copolymers are talc, chalk or glass fibers, and amounts of up to 50% by weight may be used here.

Olefin polymers preferably used for producing the injection stretch blow molded containers comprise nucleating agents 40 suitable for improving transparency. Examples of usual nucleating agents are inorganic additives, such as talc, silica or kaolin, salts of mono- or polycarboxylic acids, such as sodium benzoate or aluminum tert-butyl benzoate, or salts of phosphoric diesters, such as sodium 2,2'-methylenebis(4,6-di-tert-butylphenyl) 45 phosphate. Nucleating agents whose use is preferred are dibenzylidenesorbitol or its C₁-C₈-alkyl-substituted derivatives such as methyl- or dimethyldibenzylidenesorbitol. The content of

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nucleating agents is generally up to 5% by weight, preferably up to 3% by weight and in particular up to 1% by weight, based on the olefin polymers.

- 5 To produce the novel injection stretch blow molded containers, it is usual firstly to injection mold parisons from olefin polymers, comprising homopolymers of propylene or comprising copolymers of propylene with other C₂-C₁₀-alk-1-enes, which are obtainable by polymerizing the corresponding monomers with metallocene
- 10 catalysts, or preferably exclusively from homopolymers of propylene or copolymers of propylene with other C₂-C₁₀-alk-1-enes obtainable with metallocene catalysts. After this, the parisons are stretch blow molded.
- 15 This may take place in a single-step procedure. In that case, the injection molding of the parison and the stretch blow molding take place in the same machine, the parison being cooled rapidly to the stretching temperature. The advantage of the single-step process is in particular the saving of energy.
- 20 It is also possible to proceed in two steps. In that case, the parison is firstly injection molded. The parison is then generally allowed to cool to room temperature, making it easy to store. The parison is then heated once again for the independent
- 25 stretch blow molding step. This two-step injection stretch blow molding process is therefore also frequently termed the reheat process. An advantage of the two-step process is that the two steps are independent of one another and better control of temperature is possible during stretch blow molding.
- 30 With both versions it is possible to carry out the stretch blow molding in two steps. In that case, the parison is firstly preblown in a first step, and after that is stretch blow molded in a further step.
- 35 The injection molding of the parison usually takes place at melt temperatures of from 200 to 280°C, and the stretch blow molding at from 100 to 160°C with a stretch blow molding pressure of from 10 to 50 bar and a stretching ratio of from 1:2 to 1:7. If
- 40 preblowing is carried out, this takes place at from 110 to 150°C with a preblowing pressure of from 1 to 10 bar and a stretching ratio of from 1:2 to 1:5.

- The novel containers have, inter alia, the capability of being
- 45 produced in a wider temperature window than containers produced from olefin polymers which comprise no propylene polymers obtainable with metallocene catalysts. This permits smooth

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continuous operation with low personnel costs in manufacturing. In addition, they can be produced with reduced cycle times. The containers have uniform wall thicknesses, high gloss and excellent transparency, which may be further improved by adding
5 nucleating agents. The containers have good stiffness and toughness and, because of their low content of soluble fractions, are suitable in particular for applications in the food and drink and medical sectors.

10 Examples

Round bottles of one liter capacity and weighing 30 gram when empty were produced in an injection stretch blow molding process encompassing three steps. Firstly, a parison was injection molded
15 in a first step, and in a second step was then removed from the injection mold and transferred into a preblowing apparatus, where the parison was brought to a specified temperature and preblown at 4 bar. In a third step, the preblown parison was stretch blow molded using a pressure of 17 bar.

20

The temperature of the preblowing step was varied in each of the ranges stated, and the containers were tested for transparency and uniformity of shape. At the same time, the temperature window within which commercially acceptable products were obtainable was
25 determined.

Example 1

Propylene was homopolymerized in a gas-phase reactor with
30 vertical agitation at 60°C, at a pressure of 24 bar and with an average residence time of 1.5 h, in the presence of hydrogen as molar mass regulator, using a dimethylsilanediylbis-[3,3'-(2-methylbenzindenyl)]zirconium dichloride/methylaluminum-oxane catalyst supported on silica gel. This gave a propylene
35 homopolymer with a melting point of 147°C, an MFR (230°C/2.16 kg) of 3.8 g/10 min and an M_w/M_n ratio of 2.0.

This polymer was used to produce the abovementioned bottles by injection stretch blow molding. The temperature of the preblowing
40 step was varied in the range from 110 to 130°C. The temperature window within which highly transparent bottles of uniform wall thickness were obtained was 8°C.

Example 2

In a manner similar to that of Example 1, a propylene-1-butene copolymer was polymerized with 3.5% by weight of 1-butene. It had
5 a melting point of 134°C, an MFR (230°C/2.16 kg) of 3.8 g/10 min and an M_w/M_n ratio of 2.2.

Bottles were produced by injection stretch blow molding as in Example 1. The temperature of the preblowing step was varied in
10 the range from 110 to 130°C. The temperature window in which highly transparent bottles of uniform wall thickness were obtained was 12°C.

Comparative Example A

15

As in Example 1, bottles were produced by injection stretch blow molding using a commercial propylene homopolymer prepared using a conventional Ziegler- Natta catalyst (NOVOLEN® 1100L from BASF Aktiengesellschaft with a melting point of 164°C, an MFR
20 (230°C/2.16 kg) of 6 g/10 min, an M_w/M_n ratio of 5 and a content of 3% by weight of fractions soluble in xylene at room temperature). The temperature of the preblowing step was varied in the range from 140 to 160°C. The temperature window within which highly transparent bottles of uniform wall thickness were
25 obtained was 2°C.

Comparative Example B

As in Example 2, bottles were produced by injection stretch blow
30 molding using a commercial propylene copolymer with 2.5% by weight of ethylene prepared using a conventional Ziegler- Natta catalyst (NOVOLEN® 3200MC from BASF Aktiengesellschaft with a melting point of 145°C, an MFR (230°C/2.16 kg) of 8 g/10 min, an M_w/M_n ratio of 4.5 and a content of 6.5% by weight of fractions
35 soluble in xylene at room temperature). The temperature of the preblowing step was varied in the range from 115 to 140°C. The temperature window within which highly transparent bottles of uniform wall thickness were obtained was 6°C.

40

45

1. A process for producing injection stretch blow molded containers from olefin polymers by injection molding parisons at melt temperatures of from 200 to 280°C and then stretch blow molding the parisons at from 100 to 160°C, which comprises stretch blow molding olefin polymers comprising at least 40% by weight, based on the olefin polymers, of homopolymers of propylene or copolymers of propylene with other C₂-C₁₀ 1-alkenes, which are obtained by polymerizing the appropriate monomers with metallocene catalysts comprising, as active constituents,

A) one or more metallocene complexes of the general formula (Ic)



M is titanium, zirconium, hafnium, vanadium, niobium or tantalum, or else elements of the 3rd subgroup of the Periodic Table or of the lanthanoids,

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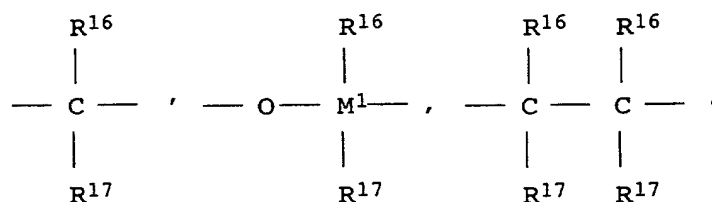
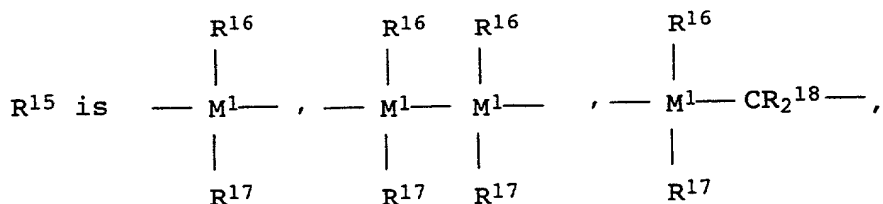
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R^6 and R^7 are C_1 - C_{10} -alkyl, C_6 - C_{15} -aryl, alkylaryl, aryl-alkyl, fluoroalkyl or fluoroaryl having in each case from 1 to 10 carbon atoms in the alkyl radical and from 6 to 20 carbon atoms in the aryl radical, and

the radicals X are identical or different,

R^1 to R^3 , R^5 , R^9 to R^{11} and R^{13} are hydrogen, C_1 - C_{10} -alkyl, 5- to 7-membered cycloalkyl, which in turn may be C_1 - C_{10} -alkyl-substituted, C_6 - C_{15} -aryl or arylalkyl, where two adjacent radicals together may also be saturated or unsaturated cyclic groups having from 4 to 15 carbon atoms, or $Si(R^8)_3$, where

R^8 may be C_1 - C_{10} -alkyl, C_3 - C_{10} -cycloalkyl or C_6 - C_{15} -aryl, and



$= BR^{16}$, $= AlR^{16}$, $-Ge-$, $-Sn-$, $-O-$, $-S-$, $= SO$, $= SO_2$,
 $= NR^{16}$, $= CO$, $= PR^{16}$ or $= P(O)R^{16}$,

where

each of R^{16} , R^{17} and R^{18} is identical or different and is hydrogen, halogen, C_1 - C_{10} -alkyl, C_1 - C_{10} -fluoroalkyl, C_6 - C_{10} -fluoroaryl, C_6 - C_{10} -aryl, C_1 - C_{10} -alkoxy, C_2 - C_{10} -alkenyl, C_7 - C_{40} -arylalkyl, C_8 - C_{40} -arylalkenyl or C_7 - C_{40} -alkylaryl, or where two adjacent radicals, in each case with the atoms linking them, form a saturated or unsaturated ring having from 4 to 15 carbon atoms, and

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M¹ is silicon, germanium or tin,

and two adjacent radicals R² and R³ and two adjacent radicals R¹⁰ and R¹¹ are in each case together a ring having from 4 to 15 carbon atoms,

and

B) one or more metallocenium-ion-forming compounds.

10

2. A process for producing injection stretch blow molded containers as claimed in claim 1, where the olefin polymers are homopolymers of propylene or copolymers of propylene with other C₂-C₁₀ 1-alkenes, obtainable by polymerizing the appropriate monomers with metallocene catalysts.

15

3. A process for producing injection stretch blow molded containers as claimed in claim 1 or 2, where the homopolymers of propylene or copolymers of propylene with other C₂-C₁₀ 1-alkenes, obtainable using metallocene catalysts, have a melting point of from 120 to 165°C.

20

4. A process for producing injection stretch blow molded containers as claimed in claims 1 to 3, where the homopolymers of propylene or copolymers of propylene with other C₂-C₁₀ 1-alkenes, obtainable using metallocene catalysts, have a melt flow rate MFR of from 2 to 50 g/10 min at 230°C with a load of 2.16 kg.

25

5. A process for producing injection stretch blow molded containers as claimed in any of claims 1 to 4, where the olefin polymers comprise up to 5% by weight, based on the olefin polymers, of nucleating agents as a further component.

30

6. An injection stretch blow molded container as claimed in any of claims 1 to 5, where the metallocene complex (Ic) is dimethylsilanedilybis[3,3'-(2-methylbenzindenyl)]zirconium dichloride.

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Injection stretch blow molded containers made from olefin polymers

5 Abstract

Injection stretch blow molded containers are obtainable from olefin polymers, comprising homopolymers of propylene or copolymers of propylene with other C₂-C₁₀-alk-1-enes, which are
10 obtainable by polymerizing the corresponding monomers with metallocene catalysts.

Processes for producing injection stretch blow molded containers from olefin polymers, and also the use of olefin polymers for
15 producing injection stretch blow molded containers, are described.

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Declaration, Power of Attorney

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We (I), the undersigned inventor(s), hereby declare(s) that:

My residence, post office address and citizenship are as stated below next to my name,

We (I) believe that we are (I am) the original, first, and joint (sole) inventor(s) of the subject matter which is claimed and for which a patent is sought on the invention entitled

Injection stretch blow molded containers made from olefin polymers

the specification of which

☒ is attached hereto.

☐ was filed on _____ as

Application Serial No. _____

and amended on _____.

☒ was filed as PCT international application

Number PCT/EP 99/00578

on 29/01/1999

and was amended under PCT Article 19

on _____ (if applicable).

We (I) hereby state that we (I) have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We (I) acknowledge the duty to disclose information known to be material to the patentability of this application as defined in Section 1.56 of Title 37 Code of Federal Regulations.

We (I) hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed. Prior Foreign Application(s)

Application No.	Country	Day/Month/Year	Priority Claimed
19805329.0	Germany	11 February 1998	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No

We (I) hereby claim the benefit under Title 35, United States Codes, § 119(e) of any United States provisional application(s) listed below.

(Application Number)

(Filing Date)

(Application Number)

(Filing Date)

We (I) hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

Application Serial No.

Filing Date

Status (pending, patented,
abandoned)

_____	_____	_____
_____	_____	_____
_____	_____	_____
_____	_____	_____

2 And we (I) hereby appoint **Messrs. HERBERT. B. KEIL**, Registration Number 18,967; and **RUSSEL E. WEINKAUF**, Registration Number 18,495; the address of both being Messrs. Keil & Weinkauff, 1101 Connecticut Ave., N.W., Washington, D.C. 20036 (telephone 202-659-0100), our attorneys, with full power of substitution and revocation, to prosecute this application, to make alterations and amendments therein, to sign the drawings, to receive the patent, and to transact all business in the Patent Office connected therewith.

We (I) declare that all statements made herein of our (my) own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Declaration

Page 3 of 4

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4-00 David Fischer
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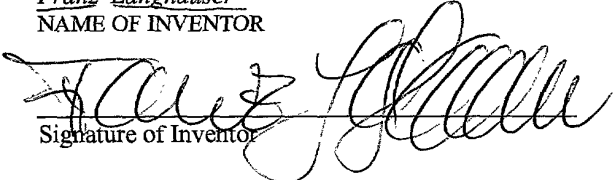
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Declaration

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0050/048773

5-00 Franz Langhauser
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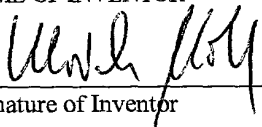

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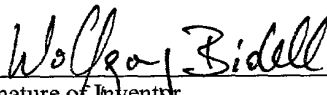

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